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Long-Term Recovery of PCB-Contaminated Surface Sediments at the Sangamo-Weston/ Twelvemile Creek/Lake Hartwell Superfund Site

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Natural recovery of contaminated sediments relies on burial of contaminated sediments with increasingly clean sediments over time (i e , natural capping) Natural capping reduces the risk of resuspension of contaminated surface sediments, and it reduces the potential for contaminant transport into the food chain by limiting bioturbation of contaminated surface or near-surface sediments This study evaluated the natural recovery of surface sediments contaminated with polychlorinated biphenyls (PCBs) at the Sangamo-Weston/Twelvemile Creek/Lake Hartwell Superfund Site (Lake Hartwell), Pickens County, SC The primary focus was on sediment recovery resulting from natural capping processes Total PCB (t-PCB), lead-210 (^{210}Pb), and cesium-137 (^{137}Cs) sediment core profiles were used to establish vertical t-PCB concentration profiles, age date sediments, and determine surface sedimentation and surface sediment recovery rates in 18 cores collected along 10 transects Four upgradient transects in the headwaters of Lake Hartwell were impacted by historical sediment releases from three upgradient sediment impoundments These transects were characterized by silt/clay and sand layering The highest PCB concentrations were associated with silt/clay layers (1 8–3 5% total organic carbon (TOC)), while sand layers (0 05–0 32% TOC) contained much lower PCB concentrations The historical sediment releases resulted in substantial burial of PCB-contaminated sediment in the vicinity of these four cores, each core contained less than 1 mg/kg t-PCBs in the surface sand layers Cores collected from six downgradient Lake Hartwell transects consisted primarily of silt and clay (0 91–5 1% TOC) and were less noticeably impacted by the release of sand from the impoundments Vertical t-PCB concentration profiles in these cores began with relatively low PCB concentrations at the sediment–water interface and increased in concentration with depth until maximum PCB concentrations were measured at ~30–60 cm below the sediment–water interface, ca 1960–1980 Maximum t-PCB concentrations were followed by progressively decreasing concentrations with depth until the t-PCB concentrations approached the detection limit, where sediments were likely deposited before the onset of PCB use at the Sangamo-Weston plant The sediments containing the maximum PCB concentrations are associated with the period of maximum PCB release into the watershed Sedimentation rates averaged 2 1 ± 1 5 g/(cm^2 yr) for 12 of 18 cores collected The 1994 Record of Decision cleanup requirement is 1 0 mg/kg, two more goals (0 4 and 0 05 mg/kg t-PCBs) also were identified Average surface sedimentation requirements to meet the three goals were 1 4 ± 3 7, 11 ± 4 2, and 33 ± 11 cm, respectively Using the age dating results, the average recovery dates to meet these goals were 2000 6 ± 2 7, 2007 4 ± 3 5, and 2022 7 ± 11 yr, respectively (The 95% prediction limits for these values also are provided) Despite the reduction in surface sediment PCB concentrations, PCB concentrations measured in largemouth bass and hybrid bass filets continue to exceed the 2 0 mg/kg FDA fish tolerance level

Introduction

Natural recovery of contaminated sediments relies on two primary mechanisms burial of contaminated sediments with clean sediments (i e , natural capping) and transformation or removal of contaminants via contaminant weathering (*1*) Natural capping reduces the risk of resuspension of contaminated surface sediments (*2*), and it reduces the potential for contaminant transport into the food chain by limiting bioturbation and bioaccumulation in surface or near-surface sediments Natural capping is a relatively slow process that relies on the deposition of increasingly lesser-contaminated sediments This occurs in net depositional environments where the rate of sediment deposition exceeds the rate of sediment scouring and resuspension (*1*) However, depositional environments also are likely repositories for contaminated sediment particles Thus, if source attenuation is not adequately addressed, the same sediment transport mechanisms that contribute to surface sediment recovery may also contribute to ongoing deposition and accumulation of contaminated particles Therefore, source removal should be an integral part of the natural recovery process if depositional environments are to result in reduced surface sediment concentrations Even after the primary source is controlled, however, recovery of surface sediments tends to be gradual due to residual or secondary contaminant sources, sediment transport processes, and benthic mixing (*3–5*)

The National Risk Management Research Laboratory (NRMRL) of the U S Environmental Protection Agency (U S EPA) is interested in developing effective, inexpensive remediation technologies for contaminated sediments, including natural recovery This study evaluated the recovery of surface sediments contaminated with polychlorinated biphenyls (PCBs) at the Sangamo-Weston/Twelvemile Creek/Lake Hartwell Superfund Site (Lake Hartwell) in Pickens County, SC The primary focus was on the recovery of PCB-contaminated surface sediments resulting from natural sedimentation

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Site Description

Lake Hartwell is an artificial lake located in northwest South Carolina, along the Georgia state line It is bordered by Anderson, Pickens, and Oconee counties in South Carolina and by Stephens, Franklin, and Hart Counties in Georgia It was created in 1955 when the U S Army Corps of Engineers (USACE) constructed Hartwell Dam on the Upper Savannah River, 11 km from its confluence with the Seneca and Tugaloo Rivers Lake Hartwell covers nearly 22 660 ha of water with a shoreline of 1500 km (*6*) The land surrounding and in close proximity to Lake Hartwell encompasses rural countryside, the City of Clemson, numerous small towns, forests, and agricultural areas

Lake Hartwell sediments were contaminated by PCBs released from the Sangamo-Weston plant, located approximately 24 km upstream along Town Creek, a tributary of Twelvemile Creek that in turn is a tributary of Lake Hartwell Sangamo-Weston manufactured capacitors from 1955 to 1978 (*7*) The plant used a variety of dielectric fluids in its manufacturing processes, including fluids containing PCBs Waste disposal practices included land burial of off-specification capacitors and wastewater treatment sludge on the plant site and at six satellite disposal areas An unspecified amount of PCBs was buried in the satellite disposal areas PCBs also were discharged with effluent directly into Town Creek Between 1955 and 1977, the average quantity of PCBs used by Sangamo-Weston ranged from 700 000 to 2 000 000 lb/yr An estimated 3% of these PCBs was discharged into Town Creek, resulting in an estimated cumulative discharge of over 400 000 lb of PCBs (*7*)

PCB use at Sangamo-Weston was terminated in 1977 (*7*) Information from the Sangamo-Weston/Twelvemile Creek/Lake Hartwell Record of Decision (*7*) and discussions with Mr Craig Zeller, the U S EPA Region 4 Site Remedial Project Manager (RPM), indicate that the Sangamo-Weston plant primarily used Aroclors 1016, 1242, and 1254 Remediation of terrestrial areas and sources has been completed (*8*)

The U S EPA's cleanup plan for Lake Hartwell relies on natural recovery, with emphasis on natural capping by the continued deposition of increasingly clean sediment entering the lake Included in the U S EPA's monitoring program are annual fish tissue and sediment monitoring, adoption of risk-based consumption guidelines for Lake Hartwell, and a public education program designed to increase awareness of fish advisories

Methods

Sediment cores were collected, segmented into 5-cm segments, and analyzed for 107 PCB congeners, lead-210 (^{210}Pb), and cesium-137 (^{137}Cs) Data were used to establish vertical total PCB (t-PCB) concentration profiles, age date sediments, and determine surface sedimentation and surface sediment recovery rates

Sediment Coring and Sample Collection Sediment cores were collected during two separate annual events conducted in the spring of 2000 and 2001 During the 2000 event, sediment cores were collected from 10 transect locations in the upper reaches of Lake Hartwell and in Twelvemile Creek, previously established by U S EPA Region 4 and USACE under the site's ongoing annual monitoring program (Figure 1) As much as possible, sample cores were taken from the deepest portion (approximately the center) of the lake at each transect A real-time global positioning system (GPS) was used to record core locations Cores from transects O, N, L, J, I, and T6 were collected in Lake Hartwell using a gravity corer off a pontoon boat, core lengths were limited by the vertical depth the gravity corer penetrated the sediment when dropped from the pontoon at water depths of 6–12 m Guide vanes on the corer ensured the collection of vertical cores

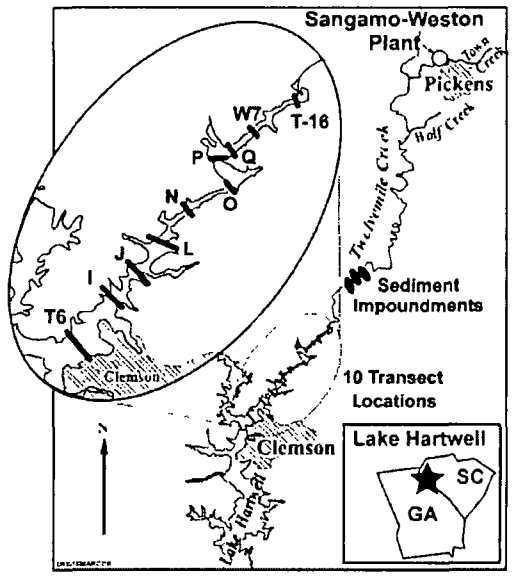

FIGURE 1. Lake Hartwell transect locations

The cores were collected in 5-cm-diameter Lexan tubes Shallower water depths of <50 cm made it possible to collect cores from Transects T16, W7, Q, and P in Twelvemile Creek on foot using a conventional sand pounder and 5-cm-diameter Lexan tubes These core lengths (135–172 cm) were limited by the ability of the field crew to pound the cores into the sediment and retract them by hand

During the 2001 sampling event, eight additional sediment cores were collected at three transect locations, all in Lake Hartwell Three cores were collected from Transect O, three from Transect L, and two from Transect I These cores were collected from shore to shore at each transect where possible, as opposed to along the deepest portion of the lake, to better understand the historical deposition and dechlorination of PCBs nearer the lake shores (Figure 2) During the 2001 sampling event, a pontoon vessel was used as the sampling platform Sediment cores were collected using a corer equipped with 7 6-cm-diameter butyrate sleeves and a mechanical vibrator to drive each core to a minimum depth of 100 cm

Different core extrusion and segmenting methods were used for the 2000 and 2001 sampling events During the 2000 sampling event, parallel cores were drawn from each location One core was dedicated for PCB analyses and one for age dating analyses Sediment samples were extruded vertically upward through the cores using a piston The water column was pushed over the top of the core tube and discarded until the sediment–water interface reached the top of the tube Then the sediment cores were extruded in 5-cm increments, each of which was collected in 8-oz glass sample jars Beginning each core segmenting process at the sediment–water interface ensured matching profiles for PCB and age dating analyses

For the 2001 sampling event, only single cores were collected at each location, the larger diameter (10 cm) cores provided sufficient sediment mass for all analyses Cores were segmented on shore into 5-cm intervals by horizontally slicing the cores with a fine-blade hacksaw Each 5-cm segment was divided into three pie segments using a small, prefabricated pie cutter that divided the segments into predetermined ratios for off-site analyses

On-site decontamination and chain-of-custody followed U S EPA-approved quality assurance project plans (QAPPs) (*9*, *10*) Core locations and sediment segments for the 2000 sampling event are identified by transect as T-T16, T-W7,

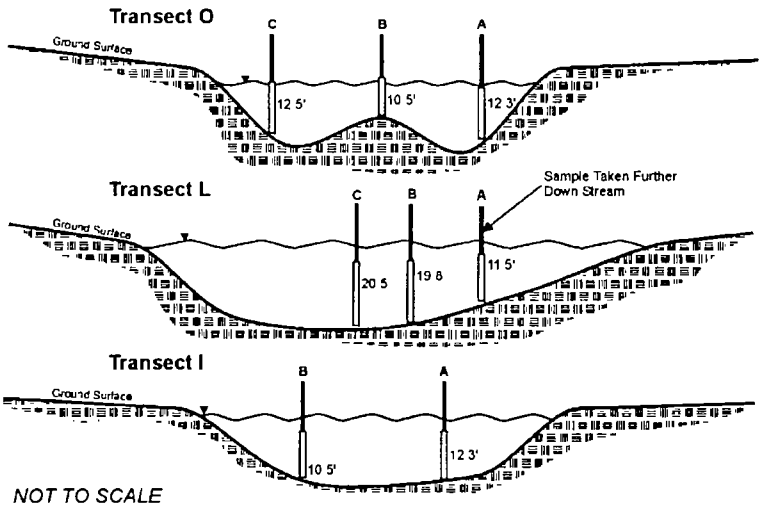

FIGURE 2. 2001 cross-transect coring locations at T-O, T-L, and T-I (Cross sections are based on visual observation and field notes)

T-Q, T-P, T-O, T-N, T-L, T-J, T-I, and T-T6 (Figure 1) For the 2001 sampling event, cross transects within T-O, T-L, and T-I are labeled T-OA, T-OB, and T-OC, T-LA, T-LB, and T-LC, and T-IA and T-IB, respectively (Figure 2) Sediment segments are identified by depth beginning at the sediment–water interface

Sediment Age Dating Sediment dating was conducted using ^{210}Pb and ^{137}Cs isotopes, two isotopes relatively common in sediments that can be used to determine the age of sediments over years or decades (*11–16*) ^{210}Pb and ^{137}Cs isotopes were analyzed at the Battelle Marine Science Laboratories (Sequim, WA) in accordance with methods described previously (*17*)

Because these two isotopes adsorb to particle surfaces, their sorbed concentrations in sediment depend on the sediment particle size distribution (PSD), e g , fine-grained sediment generally contains higher activity than sandy sediment Thus, the ability to accurately age date sediments is influenced by the uniformity of the PSD through the dated vertical profile and the historical consistency of the sedimentation rate over the dated period When these two conditions were not met (i e , there had not been a constant sedimentation rate or grain size was not uniform), the accuracy of the dating procedure was compromised To the extent possible, these conditions were confirmed visually during the initial inspection of the sediment cores and through PSD, moisture content, and ^{210}Pb vertical concentration profiles in the cores For this study, moisture content and ^{210}Pb and ^{137}Cs concentrations were determined for all core segments PSD and total organic carbon (TOC) analyses were conducted on three or more sediment segments per core for the cores collected in 2000, and on all segments for those collected in 2001

Sediment PCB Analyses Sediment PCB analyses were conducted at the Battelle Ocean Science Laboratories (Duxbury, MA) Homogenized sediments (approximately 30-g wet mass) were extracted and analyzed using modified U S EPA SW-846 Method 8270 Each sample was spiked with surrogate internal standards (PCB14, PCB34, PCB104, and PCB112) before extraction to monitor extraction efficiency Congener concentrations were not surrogate corrected, recoveries averaged 80 ± 49% for 2000 and 65 ± 16% for 2001, for all four compounds and all samples (see the Supporting Information for surrogate recovery details)

The sediment samples were extracted three times using 100 mL of hexane for each extraction, for 12, 4, and 1 h, respectively After each extraction, samples were centrifuged, solvent decanted, and concentrated to ~1 mL The hexane extract was solvent exchanged into methylene chloride and then processed through a 20-g alumina column (2% deactivated F20) to purify the PCB extract The concentrated extract was treated with activated granular copper to complex sulfur, and further purified using high-performance liquid chromatography/gel permeation chromatography (HPLC/GPC) to remove coextracted biogenic materials Final extracts were spiked with appropriate concentrations of recovery internal standard (PCB36, PCB96, PCB103, and PCB106) for low-resolution gas chromatography/mass spectrometry (GC/MS) analysis

Sediment extracts were analyzed for the concentration of 107 PCB congeners using modified Method 8270 (see the Supporting Information for the list of congeners) The analytical system was comprised of a Hewlett-Packard (HP) 6890 gas chromatograph, equipped with an electronic pressure-controlled (EPC) inlet and an HP 5973 MSD operating in the selected ion monitoring (SIM) mode Five-point standards ranged from ~0 05 to ~1 ng/μL, and samples were bracketed by analyzing standard checks no less frequently than every 10 samples and at the completion of each sample sequence The t-PCB concentration was determined as the sum of the individual PCB congeners

TOC, PSD, and Moisture Content Analyses A total carbon analyzer (UIC, Inc , Chicago, IL) was used to determine sediment TOC in accordance with U S EPA Method 9060-Total Organic Carbon PSD was performed by Soil Technology Inc (Bainbridge, WA) according to the American Society for Testing and Materials (ASTM) D422 (*18*) PSD data were reported as weight percentages of gravel (>4 74-mm diameter), sand (2–0 0625-mm diameter), silt (0 0625-mm to 4-μm diameter), and clay (<4-μm diameter) PSD analyses for cores T-J, T-P, T-Q, T-W7, and T-T16 indicated nonuniform historical sediment deposition, so these cores could not be age dated Moisture content was determined gravimetrically using ASTM Method D2216 (*19*)

Results and Discussion

Vertical t-PCB Concentration Profiles Vertical sediment profiles for the site for the 2000 sampling period were divided into two regions The four most upgradient Transects (T-T16, T-W7, T-Q, and T-P) resided in Twelvemile Creek at the headwaters of Lake Hartwell and were impacted significantly by historical sediment releases from behind three upgradient sediment impoundments These impoundments were used to generate electricity and were flushed periodically to release sediments into the lake environment Transects

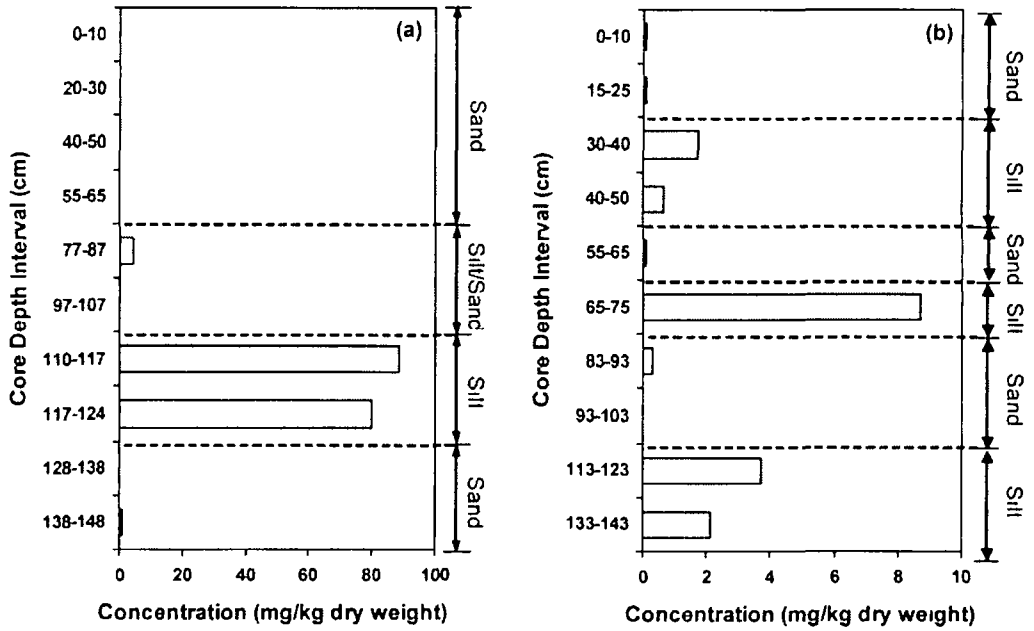

FIGURE 3 Vertical t-PCB concentration profiles for upgradient Transects T-T6 (a) and T-Q (b) Silt/sand layers are shown on the right-hand side of each figure Vertical axes are not to scale (Profiles for Transects T-W7 and T-P are shown in the Supporting Information.)

TABLE 1. Surface Sediment t-PCB Concentrations and Maximum t-PCB Concentrations Measured in Twelvemile Creek Cores T-T16, T-W7, T-Q, and T-P (Collected in 2000)

core	surface sand layer depth (cm)	t-PCB concn range in the surface sand layers of each upgradient core (mg/kg)	max core t-PCB concn (mg/kg) (corresponding depth (cm))
T16	65	0 014–0 090	88 7 (110–117)
W7	91	0 024–0 977	21 7 (94–104)
Q	25	0 076–0 092	8 71 (65–75)
P	80	0 066–0 923	138 (95–105)

T-O, T-N, T-L, T-J, T-T6, and T-I resided further downgradient in Lake Hartwell, cores from these transects consisted primarily of silt/clay and were less noticeably impacted by sand releases from the impoundments

Figure 3 shows t-PCB concentrations plotted against core depth for two of the upgradient cores collected at Transects T-16 and T-Q In both cores, the highest PCB concentrations were associated with silt/clay layers, which contained 57 ± 6 9% silt and clay and 2 6 ± 0 86% TOC The sand layers contained 96 ± 2 9% sand and 0 21 ± 0 08% TOC and had much lower PCB concentrations Vertical t-PCB profiles for Transects T-W7 and T-P resembled the profiles for Transects T-16 and T-Q (see the Supporting Information)

The historical sediment releases resulted in substantial burial of PCB-contaminated sediment by sand with low t-PCB concentrations (Table 1), so that each upgradient core contained less than 1 mg/kg t-PCBs in the surface sand layers, thus meeting the surface sediment cleanup goal of 1 mg/kg for Lake Hartwell (7) Much higher PCB concentrations were buried (Table 1), and the maximum t-PCB concentrations in these cores ranged from 8 71 to 138 mg/kg

Cores collected in 2000 from Transects T-O, T-N, T-L, T-J, T-T6, and T-I consisted primarily of silt and were not noticeably impacted by the release of silt and sand from the impoundments Sediment t-PCB concentrations are plotted against depth in Figure 4 for all six Lake Hartwell cores, each figure also shows the 1 0 mg/kg cleanup goal Of these six downgradient cores, only cores from Transects T-L (Figure 4c) and T-T6 (Figure 4f) fully penetrated the PCB-contaminated sediments to provide a complete vertical profile of PCB contamination For T-L, the vertical t-PCB concentration profile began with relatively low PCB concentrations at the sediment–water interface and increased in concentration with depth until the maximum PCB concentration was measured at the 35–40-cm interval The maximum t-PCB concentration was followed by progressively decreasing concentrations with depth until the t-PCB concentration approached the detection limit at ~85 cm below the sediment–water interface, where sediments were likely deposited at the onset of PCB use at Sangamo-Weston The sediments containing the maximum PCB concentrations are associated with the period of maximum PCB release into the watershed For Transect T-T6, the maximum concentration occurred at the 15–20-cm depth interval, the shallower depth may be explained by the observation that Transect T-T6 had the lowest measured sedimentation rate (see age dating below) The behavior of the core T-J profile was inconsistent with that of the other cores collected in 2000 and cannot readily be explained, core T-J was represented by relatively low t-PCB concentrations (~1–1 5 mg/kg) over the entire 30-cm profile

Sediment t-PCB concentrations (mg/kg) are plotted against depth in Figure 5 for the Lake Hartwell cores collected in 2001 at Transects T-O (Figure 5a–c), T-L (Figure 5d–f), and T-I (Figure 5g,h) The more aggressive coring method employed in 2001 ensured that all cores recovered at least 100 cm, providing more complete concentration profiles than those obtained from the 2000 cores Core T-OB also recovered 100 cm, but was analyzed only to a depth of 65 cm, below which sand levels ranged from 86% to 93%

The highest buried t-PCB concentrations occurred at Transect L and ranged from 14 1 mg/kg at the 30–35-cm interval in core T-LA to 66 4 mg/kg at the 30–35-cm interval in core T-LC Maximum t-PCB concentrations in the Transect O cores were less than 16 5 mg/kg The maximum t-PCB concentrations at Transect I were 19 3 and 25 1 mg/kg and occurred in the 15–20- and 20–25-cm intervals in cores T-IA and T-IB, respectively The shallower PCB deposition at Transect I compared to Transects L and O is consistent with the fact that the T-I cores had the lowest sediment ac-

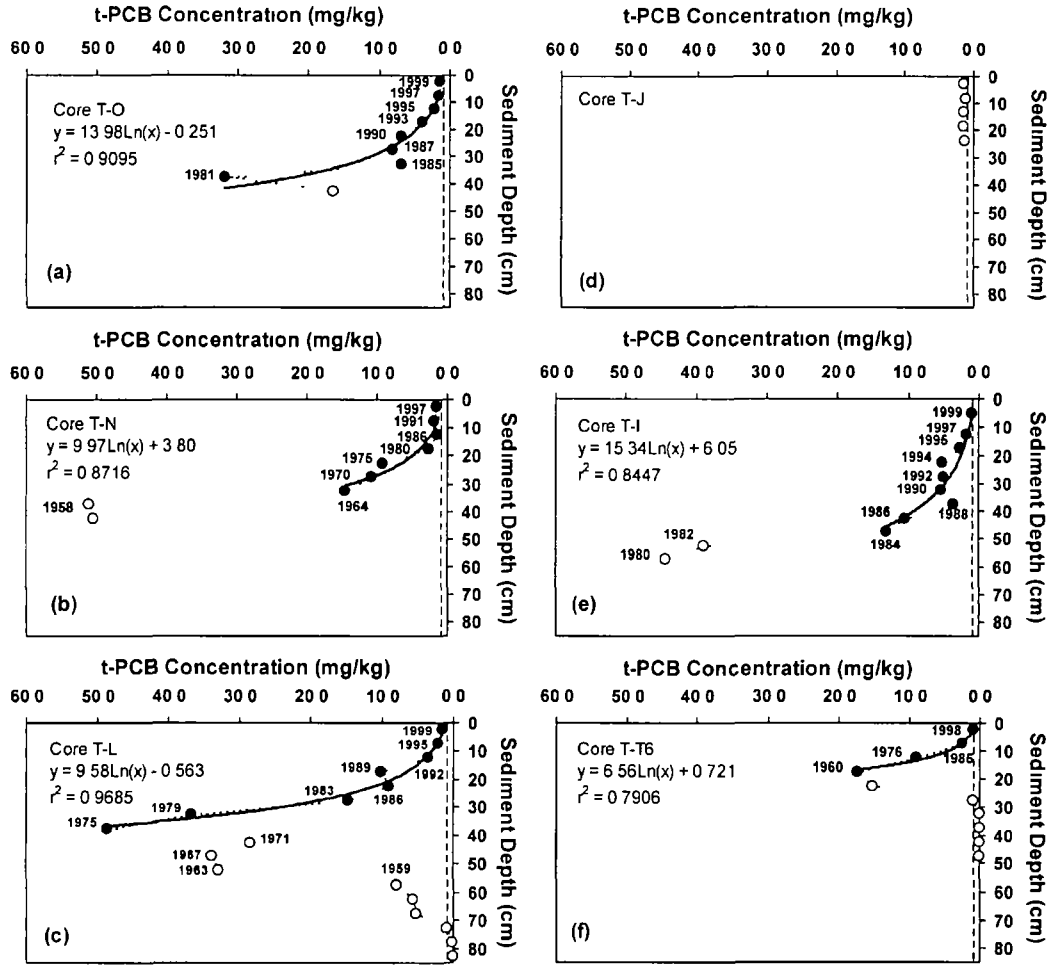

FIGURE 4 Sediment t-PCB concentration (mg/kg) plotted against depth below the sediment–water interface for cores T-O, T-L, T-I, T-N, T-J, and T-T6 collected in 2000 Best-fit logarithmic curves are shown with corresponding equations and correlation coefficients to estimate surface sediment recovery. Solid symbols (●) represent data used to generate the curves Vertical dashed lines represent the 1 mg/kg cleanup goal

cumulation rates (see age dating below) for the 2001 cores

There were two patterns observed in the 2001 cores that were not seen in the 2000 cores The first relates to core T-OB, which had a relatively low maximum t-PCB concentration (8 79 mg/kg, Table 2) compared to the other cores, and more importantly exhibited an inverted t-PCB concentration profile where the highest t-PCB concentration was measured near the sediment–water interface and concentrations decreased with depth The t-PCB concentration profile was likely influenced by the unique sand distribution in this core, which had an average of 61 ± 21% sand and 5 0 ± 1 9% TOC in the upper 50 cm, and 83 ± 5 8% sand and 0 63 ± 0 69% TOC in the bottom 50 cm Sand contents in cores T-OA (45 ± 16%) and T-OC (52 ± 20%) also were relatively high compared to those in the Transect L cores at T-LA (13 ± 13%), T-LB (20 ± 16%), and T-LC (18 ± 19%), for T-LA, below 50 cm, sand increased to 77% and t-PCB concentrations decreased accordingly Average sand contents in the upper 35 cm of cores T-IA and T-IB (where PCBs were detected) were 22 ± 18% and 23 ± 21%, respectively, below 35 cm, sand in core T-IA remained relatively uniform, but sand in core T-IB increased to 44 ± 20%

Two factors likely influenced the higher sand content in the Transect O cores First, Transect O is located furthest upgradient, closer to the headwaters of Lake Hartwell, and thus is more significantly influenced by sand released from the sediment impoundments in Twelvemile Creek Second, Transect O resides in a relatively narrow channel of Lake Hartwell where flow velocities would support higher shear velocities and preferential sand deposition Cores T-LA and T-IA also had relatively high sand levels below ~35–50 cm, which influenced the distribution of t-PCB concentrations with time

The second unusual observation in the 2001 cores was the intermittent t-PCB concentration loading with depth seen primarily in cores T-OA (Figure 5a), T-OC (Figure 5c), and T-LC (Figure 5f) The low t-PCB concentrations measured at the 50–65-cm interval in core T-LC, the 40–45- and 55–60-cm intervals in core T-OA, and the 85–90-cm interval in core T-OC represent intermittent loadings of relatively clean sediment, possibly influenced by external circumstances such as storm events or regional development

Surface sediment concentrations of all 2000 cores approached the 1 0 mg/kg cleanup goal in the surface 5 cm, and in the case of Transect T6, the t-PCB concentration was slightly below 1 0 mg/kg (Table 2) Maximum t-PCB concentrations were measured around 35–40 cm depth, except for cores T-I and T-T6, where the maximum values were observed at 55–60 and 15–20 cm, respectively The lake widens significantly immediately upgradient of T-T6, compared to the relatively narrow channel that comprises the other transects, which may have influenced the shallower deposition in the vicinity of T-T6 This conclusion is supported by the age dating results, which indicate that T-T6

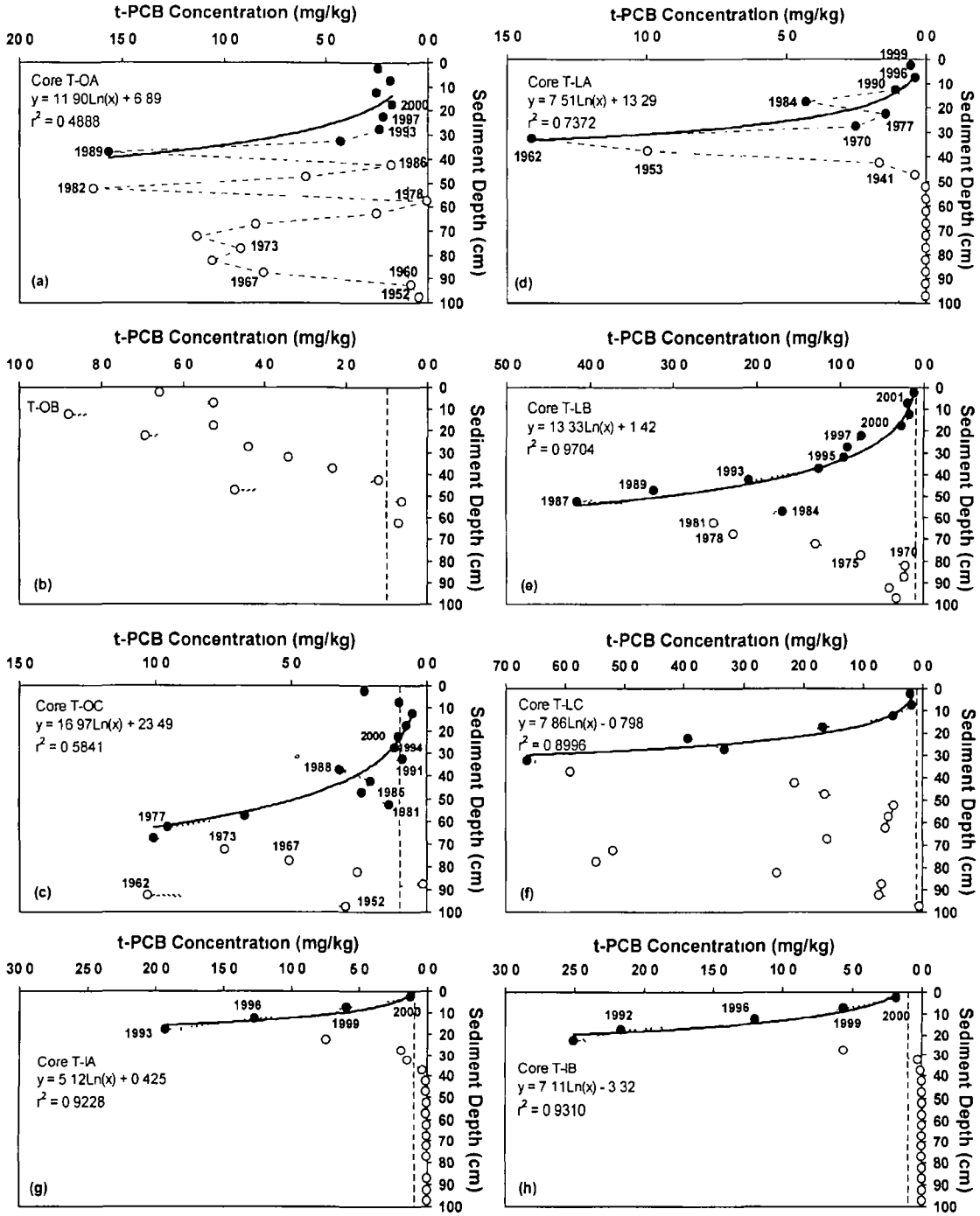

FIGURE 5. Sediment t-PCB concentration (mg/kg) plotted against depth below the sediment–water interface for cores T-LA, T-LB, T-LC, T-OA, T-OB, T-OC, T-IA, and T-IB collected in 2001. Best-fit logarithmic curves are shown with corresponding equations and correlation coefficients to estimate surface sediment recovery Solid symbols (●) represent data used to generate the curves. Vertical dashed lines represent the 1 mg/kg cleanup goal

had the lowest sedimentation rate measured in this study (see age dating below)

The 2001 surface sediment concentrations were somewhat higher, on average, than those collected in 2000, particularly at Transect O, where surface concentrations appeared to plateau in the upper 30 cm in core T-OA (Table 2, Figure 5) The surface 5-cm t-PCB concentrations averaged 1 37 ± 0 26 mg/kg in 2000 compared to 2 27 ± 1 86 mg/kg in 2001, and the surface 10-cm t-PCB concentrations averaged 1 67 ± 0 63 mg/kg in 2000 compared to 3 07 ± 2 21 mg/kg in 2001 The higher concentrations in 2001 may be attributed partially to the proximity of the cores to the shore, where less longitudinal deposition and recovery may have occurred or where more hydrodynamic mixing could have occurred Drought conditions since ~1988 also may have influenced near shore surface sediment mixing and transport by exposing lakeshore and contaminated surface sediments (observed visually), making exposed sediments more susceptible to sloughing and runoff, however, this hypothesis could not be supported by the existing data

Sediment Age Dating Sediment age dating was conducting using ^{210}Pb and ^{137}Cs profiles in the sediment cores for

TABLE 2 Surface Sediment t-PCB and Maximum t-PCB Concentrations Measured in Lake Hartwell Cores T-O, T-N, T-L, T-J, T-I, and T-T6 (Collected in 2000) and Lateral Transect Cores O-A, O-B, O-C, L-A, L-B, L-C, I-A, and I-B (Collected in 2001)

transect core	surface t-PCB concn (mg/kg) 0–5 cm	5–10 cm	max core concn[a] (mg/kg)	depth interval (cm), age date corresponding to peak t-PCB concn
		2000 Transect Cores		
O	1 38	1 62	31 9	35–40, ca 1981
N	1 50	1 81	51 1	35–40, ca 1958
L	1 58	2 12	48 7	35–40, ca 1975
J[b]	1 45	1 09		
I[c]	0 858		44 4	55–60, ca 1980
T6	0 930	2 55	17 4	15–20, ca 1960
		2001 Transect Cores		
O-A	2 44	1 85	16 4	50–55, ca 1982
O-B	6 59	8 79	8 79	10–15
O-C	2 27	1 01	10 1	60–65, ca 1977
L-A	0 55	0 39	14 1	30–35, ca 1962
L-B	1 15	1 91	41 5	50–55, ca 1987
L-C	2 07	1 78	66 4	30–35
I-A	1 23	5 90	19 3	15–20, ca 1993
I-B	1 86	5 64	25 1	20–25, ca 1992

[a] During 2000, only cores from T-L and T-T6 fully penetrated the PCB-contaminated sediments to show a complete profile of PCB contamination Therefore, it is uncertain whether the maximum t-PCB concentrations measured in cores T-O, T-N, T-J, and T-I represent true maxima [b] T-J extended only to a depth of 27 cm, where peak t-PCB concentrations were not apparent [c] The surface T-I interval spanned 0–10 cm

the 2000 and 2001 sampling events Cores with PSD analyses that revealed nonuniform historical sediment deposition could not be dated using the ^{210}Pb data The sediment release events from the upgradient impoundments in Twelvemile Creek created multiple silt/sand layers in the river and lake, which were most evident in Transects T-T16, T-W7, T-Q, and T-P (Figure 3), and which made it impossible to age date these cores The ^{210}Pb profiles also did not support age dating for the Transect T-J (Figure 4d) and T-LC (Figure 5f) cores Several cores could be dated only in the surface sediments (Figures 4 and 5) because of nonuniform deposition and silt/sand heterogeneity in deeper sediments A somewhat unconventional approach was used to age date several of the 2001 cores where segments with high sand contents were removed from the age dating analysis on the assumption that the high-sand layers were likely caused by short-term releases, this analysis was supported by calibrating the ^{210}Pb age dating profiles to ca 1955 using ^{137}Cs

The downgradient Lake Hartwell cores that were age dated included five cores from 2000 (Figure 4) and six cores from 2001 (Figure 5) In 2000, the maximum t-PCB concentrations appeared ca 1960–1980, between approximately 35 and 55 cm below the sediment–water interface Maximum t-PCB concentrations at Transect L occurred ca 1975 in core T-L, 1962 in core T-LA, and 1987 in core T-LB In core T-OA, two peaks were measured ca 1952 and 1973, and in core T-OC a peak occurred ~10 cm prior to sediments dated ca 1952 Sedimentation dates that precede 1960 suggest that these sediments represent the very early formation of the lake, which was constructed in 1955 Because discharge of PCBs into the lake began ca 1955, pre-1955 peaks in T-OA and T-OC may suggest downward porewater migration of PCBs, or they may be influenced by the relative accuracy of the age dating process

Transect O, the most upgradient transect, had the highest sedimentation rates, followed by Transect L and then Transect I (Table 3) At Transect I, the maximum concentration for

TABLE 3. Sedimentation Rates in Cores along the Centerline Transect of Lake Hartwell (2000) and along the Banks of Lake Hartwell at Transects O, L, and I (2001)

sedimentation rates in 2000 cores		sedimentation rates in 2001 cores	
transect core	sedimentation rate (g/(cm^2 yr))	transect core	sedimentation rate (g/(cm^2 yr))
O	3 45	O-A	2 46
N	0 52	O-B	5 50
L	0 77	O-C	2 73
I	1 18	L-A	1 05
T6	0 23	L-B	3 26
av ± std dev	1 23 ± 1 29	I-A	2 02
		I-B	2 21
		av ± std dev	2 75 ± 1 39

the 2000 cores occurred in the early 1980s (Figure 4e) and for the 2001 cores in the early 1990s (Figure 5g,h) The Transect I peaks in 2001 were shallower than those at Transects O and L due to the lower sedimentation rate in the vicinity of Transect I and the longer travel time for sediments to reach this transect, which is further downgradient from the headwaters of the lake than Transects L and O The lowest sedimentation rate of 0 23 mg/(cm^2 yr) was measured in core T-T6, located furthest downgradient in the lake

Surface Sediment Recovery Rates Using the surface sedimentation requirements and age dating results, the time for surface sediment recovery was predicted for each transect to achieve the cleanup goals stipulated in the ROD These goals include the 1 0 mg/kg sediment cleanup requirement and the two more restrictive goals of 0 4 and 0 05 mg/kg t-PCBs (*7*) The ROD reported that the 0 4 and 0 05 mg/kg goals were established on the basis of the National Oceanic and Atmospheric Administration's effects range-median and effects range-low published criteria associated with biological effects on aquatic life

To estimate the rate of surface sediment recovery, best-fit logarithmic regressions were fitted to the surface sediment data (Figures 4 and 5, logarithmic equations and correlation coefficients are shown with each figure) Best-fit logarithmic regressions were calculated to determine 95% prediction intervals for the dates when cleanup goals would be met Using these equations, sedimentation requirements to achieve each of the surface sediment cleanup goals were determined by setting the t-PCB value (i e , the *x*-axis value) to 1 0, 0 4, or 0 05 mg/kg, respectively (Table 4)

Very little sedimentation was required to achieve the 1 0 mg/kg goal, negative values in Table 4 mean that the equations used to determine surface sediment concentrations estimated that the 1 0 mg/kg target concentration had already been achieved, as was the case for cores T-I, T-T6, T-LA, and T-OC (Table 2) Average sedimentation requirements to achieve the 1 0 mg/kg target were higher for the 2000 cores than for the 2001 cores, however, the 2001 depths were influenced by the results of cores T-OC and T-LA, which achieved the 1 0 mg/kg goal at 20–25 and 15–20 cm below the sediment–water interface, respectively (Figure 5) T-OC appeared to rebound above 1 mg/kg in the upper 10 cm, but this had little impact on the trajectory of the best-fit curve Excluding cores T-OC and T-LA, the average 2001 sedimentation requirement (1 6 ± 3 8 cm) was closer to the average 2000 value (Table 4, footnote *f*) As expected, much greater sedimentation was required to achieve the 0 4 and 0 05 mg/kg goals Once again, the results for cores T-OC and T-LA influenced the 2001 results significantly, sedimentation requirements excluding these two cores were comparable to the depths estimated using the 2000 cores (Table 5, footnote *f*)

TABLE 4. Required Sedimentation To Achieve 1.0 mg/kg,[a] 0.4 mg/kg,[b] and 0.05 mg/kg[c] t-PCB Concentrations in the Upper 5-cm Surface Sediments[d,e]

transect core	sedimentation required to achieve 1 mg/kg t-PCB (cm)	sedimentation required to achieve 0 4 mg/kg t-PCB (cm)	sedimentation required to achieve 0 05 mg/kg t-PCB (cm)
		2000 Transect Cores	
O	2 8 (14 5)	15 6 (30 3)	44 6 (68 4)
N	−1 3 (11 8)	7 8 (23 2)	28 6 (51 0)
L	3 1 (10 2)	11 8 (19 9)	31 8 (42 5)
I	−3 5 (13 4)	10 5 (30 6)	42 4 (72 1)
T6	1 8 (30 1)	7 9 (44 0)	21 4 (77 4)
av ± std dev	0 55 ± 2 9 (16 0 ± 8 0)	11 7 ± 3 2 (29 6 ± 9 3)	33 7 ± 9 7 (62 3 ± 14 8)
		2001 Transect Cores	
OA	−4 4 (23 6)	6 5 (41 2)	31 3 (86 8)
OC	−20 0 (11 2)	−5 4 (29 3)	29 8 (75 6)
LA	−10 7 (−1 1)	−3 9 (14 4)	11 7 (36 5)
LB	1 1 (9 0)	13 3 (22 0)	41 0 (52 3)
LC	3 3 (16 2)	10 5 (24 8)	26 9 (45 2)
IA	2 1 (15 6)	6 8 (23 1)	17 4 (41 7)
IB	5 8 (17 3)	12 3 (26 3)	27 1 (47 4)
av ± std dev[f]	−3 4 ± 9 5 (13 1 ± 7 8)	5 7 ± 7 6 (25 9 ± 8 2)	26 4 ± 9 5 (55 1 ± 18 8)

[a] ROD surface sediment cleanup goal (as reported by the U S EPA, 1994) [b] Mean value for site-specific sediment quality criteria calculated using the U S EPA's equilibrium partitioning approach (as reported by the U S EPA, 1994) [c] NOAA effects range-low, based on an evaluation of published criteria associated with biological effects on aquatic life (as reported by the U S EPA, 1994) [d] Negative values imply that the calculations estimate the cleanup goal is achieved beneath the existing sediment surface [e] Values in parentheses are 95% upper prediction levels [f] Average values excluding T-LA and T-OC were 1 6 ± 3 8 cm (1 mg/kg), 9 9 ± 3 1 cm (0 4 mg/kg), and 28 7 ± 8 5 cm (0 05 mg/kg)

TABLE 5 Estimated Dates That the 1.0 mg/kg,[a] 0.4 mg/kg,[b] and 0.05 mg/kg[c] t-PCB Concentration Goals Will Be Achieved[d]

transect core	estimated dates to achieve 1 mg/kg[a] t-PCB (cm)	estimated dates to achieve 0 4 mg/kg[b] t-PCB (cm)	estimated dates to achieve 0 05 mg/kg[c] t-PCB (cm)
		2000 Transect Cores	
O	2000 7 (2006 9)	2006 7 (2014 5)	2020 4 (2033 0)
N	1995 1 (2010 0)	2004 9 (2022 4)	2027 1 (2052 6)
L	2001 6 (2011 2)	2008 0 (2018 8)	2022 7 (2036 8)
I	2000 1 (2011 2)	2005 6 (2020 3)	2017 9 (2042 1)
T6	2008 2 (2042 3)	2018 2 (2062 8)	2041 0 (2111 4)
av ± std dev	2001 1 ± 4 7 (2016 3 ± 14 6)	2008 6 ± 5 4 (2027 8 ± 19 8)	2025 8 ± 11 5 (2055 2 ± 29 1)
		2001 Transect Cores	
OA	1999 6 (2015 9)	2003 2 (2023 6)	2011 4 (2044 0)
OC	1992 5 (2010 0)	1998 7 (2019 3)	2012 8 (2045 4)
LA	1987 9 (2008 9)	1996 8 (2019 5)	2016 9 (2047 6)
LB	2003 0 (2008 7)	2006 5 (2013 0)	2014 3 (2023 1)
IA	2001 6 (2011 8)	2003 8 (2016 2)	2009 0 (2027 4)
IB	2003 0 (2011 8)	2005 8 (2016 8)	2012 2 (2028 8)
av ± std dev[e]	1997 9 ± 6 3 (2011 2 ± 2 7)	2002 5 ± 3 9 (2018 1 ± 3 6)	2012 8 ± 2 7 (2036 1 ± 10 8)

[a] ROD surface sediment cleanup goal (U S EPA, 1994) [b] Mean value for site-specific sediment quality criteria calculated using the U S EPA's equilibrium partitioning approach (as reported by the U S EPA, 1994) [c] NOAA effects range-low, based on an evaluation of published criteria associated with biological effects on aquatic life (as reported by the U S EPA, 1994) [d] Values in parentheses are 95% upper prediction levels [e] Average values excluding T-LA and T-OC were 2001 8 ± 1 6 (1 mg/kg), 2004 8 ± 1 6 (0 4 mg/kg), and 2011 7 ± 2 2 (0 05 mg/kg)

Best-fit regressions also were used to estimate the time for surface sediment recovery at each coring location by plotting sediment age (*y*-axis) versus sediment concentration (*x*-axis) data Best-fit logarithmic regressions were calculated to determine 95% prediction limits (Table 5) for time to recovery (Figure 6 shows cores for Transects T-O and T-L, for examples) Estimated surface sediment recovery to 1 mg/kg occurs ca 1995−2003, except for cores T-OC and T-LA for which the estimated times occur ca 1981 and 1987, respectively The estimated times for T-OC and T-LA cores to achieve 1 mg/kg are consistent with the estimated sedimentation requirements discussed previously and shown in Figure 5 and Table 2 and the fact that t-PCB concentrations in both cores were below 1 mg/kg in the upper 10 cm Assuming the recovery of surface sediment concentrations continues to proceed asymptotically toward background, surface sediments are estimated to reach 0 4 mg/kg by 2020 and 0 05 mg/kg by 2041

Combining the sediment concentration profiles and age dating results provided valuable information pertaining to the range of time anticipated to achieve surface sediment concentration goals Nonetheless, there is an inherent challenge in making future predictions based on extrapolation of past data trends, especially when the predictions are not based on a mechanistic understanding or verifiable model of PCB fate processes For example, there is no guarantee that the PCB fate processes that caused the exponential decay of surface sediment PCB concentrations since PCB use was terminated at Sangamo Weston will continue at the same rate in the future Although the statistical analysis provided upper 95% prediction limits based on the data trends and tempers expectations of future recovery, the reader may question whether future predictions based on the last 20 yr of sediment PCB concentration decline are appropriate It may be possible, for example, that a plateau has been reached for the concentration decline, where PCBs resurfacing from depth through advective transport and sediment mixing processes are balanced with the dilution of surface sediments through natural capping by less-contaminated sediments For these reasons, long-term monitoring must remain an

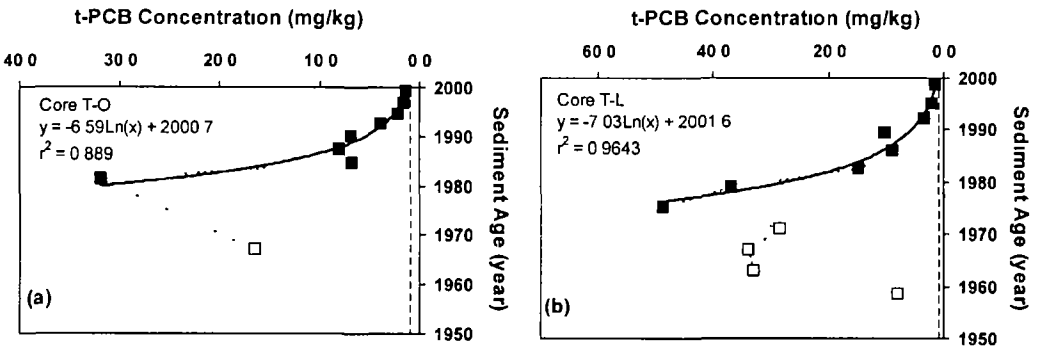

FIGURE 6 Sediment t-PCB concentration (mg/kg) plotted against sediment age for cores T-O and T-L collected in 2000. Best-fit logarithmic curves are shown with corresponding equations and correlation coefficients to estimate surface sediment recovery. Solid symbols (■) represent data used to generate best-fit curves Similar calculations were conducted for the remaining core profiles for cores T-N, T-I, T-T6, T-LA, T-LB, T-OA, T-OB, T-IA, and T-IB (Table 5). Vertical dashed lines represent the 1 mg/kg cleanup goal

integral component of the natural recovery remedy for this site Only through long-term monitoring can we determine whether the recovery of surface sediments will indeed continue into the future In addition to establishing expectations of future surface sediment recovery, this analysis also may be useful to design a long-term monitoring program to determine sampling locations, methods, frequency, and duration

The accuracy of this analysis also depends on the quality of the data, the thickness of the sediment segments, and the consistency and predictability of surface sedimentation rates For this study, cores were extruded into 5-cm intervals Narrower intervals could have provided more precise results, but the thickness of the intervals had to be weighed against the increased sampling and analysis and associated costs Mechanisms that influence the rate of surface sediment recovery include sedimentation rates, hydraulic and benthic mixing of surface sediments, and the ongoing PCB load into the lake, more rapid sedimentation of cleaner sediments will enhance recovery, while increased benthic mixing will likely retard recovery The range in the sedimentation rates, surface sedimentation requirements, prediction limits, and confidence intervals to meet the ROD cleanup goals represent the variability associated with sediment core profiling and age dating

Potential for Fish Recovery Surface sediment recovery relies on the deposition of increasingly lesser contaminated sediments in net depositional areas At Lake Hartwell, sediments were impacted historically by the release of PCB-contaminated wastewater and runoff from the former Sangamo-Weston plant and satellite disposal areas Efforts to reduce PCB levels entering the lake by removing PCB sources along with the discontinued use of PCBs since 1978 have largely met with success, as indicated by the historical recovery of surface sediments (Figures 4 and 5) Over a period of ~25 yr, a 10−50-fold reduction has been measured in the sediments, from peak concentrations of 10−66 mg/kg in buried sediments to approximately 1−3 mg/kg in surface sediments Determining whether such concentration reductions are sufficient to protect human and environmental receptors was outside of the scope of this study, but remains an important question for the site For natural recovery to be truly effective for this site, it also will be necessary to demonstrate the recovery of ecological receptors, especially fish that are consumed by recreational and subsistence anglers It is also notable that natural recovery at Lake Hartwell relies on the permanence of Hartwell dam Future removal of Hartwell dam would risk resuspension, release, and downstream migration of historically-buried, PCB-contaminated sediments

EPA Region 4 has pursued a long-term monitoring program that includes monitoring largemouth and hybrid

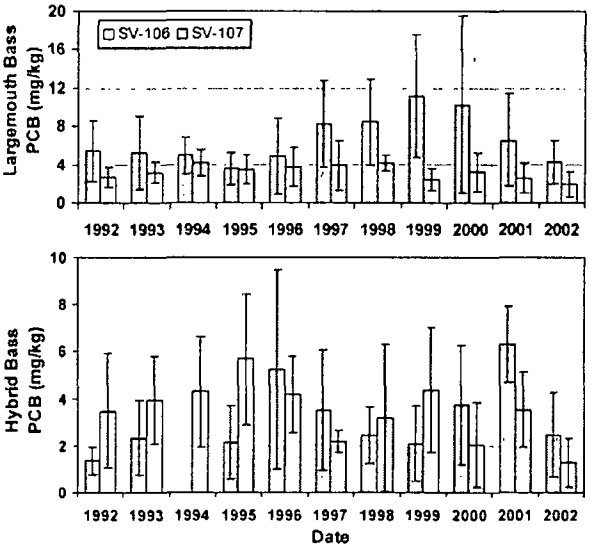

FIGURE 7 Largemouth bass and hybrid bass fish filet concentrations over time. Fish sampling was conducted by EPA Region 4 between 1995 and 2002, according to procedures described in the EPA ROD (7). Two of six locations (SV-106 and SV-107) are shown, due to their proximity to sediment coring locations

bass fish filets (*20, 21*) (The fish analyses were conducted outside of this research program and the purview of the referenced EPA QAPPs (*9, 10*) Fish were prepared for analyses in accordance with the standard FDA filet method and were analyzed by the EPA SW-846 Method 8082 PCB Aroclor method (*21*)) Ten years of monitoring between 1992 and 2002 (Figure 7) suggests that largemouth and hybrid bass fish have not measurably responded to the decreased surface sediment concentrations (the two stations, 106 and 107, were in the general vicinity of the sediment coring work (Figure 1)) These results suggest that there are other mechanisms at Lake Hartwell, in addition to surface sediment concentration, that influence fish PCB concentrations Ongoing EPA studies at Lake Hartwell are investigating these influences by measuring the potential for advective porewater PCB transport, short-term (28-day) accumulation in suspended caged fathead minnows and clams in the water column, PCB uptake into indigenous biological species at the site (e g , phytoplankton, zooplankton, macroinvertebrates, and small resident fish), and PCB volatilization into the atmosphere EPA Region 4 also maintains a public education program to heighten local awareness of the presence of PCBs in lake fish

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Supporting Information Available

Tables giving the PCB surrogate recoveries and target PCB analytes and figures showing the t-PCB vertical concentration profiles in upgradient cores T16, W7, and P This material is available free of charge via the Internet at http //pubs acs org

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